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Fast Neutron Radioactivity and Damage Studies on Materials*

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Abstract

Many materials and electronics need to be tested for the radiation environment expected at linear colliders (LCs) to improve reliability and longevity since both accelerator and detectors will be subjected to large fluences of hadrons, leptons and gammas. Examples include NdFeB magnets, considered for the damping rings, injection and extraction lines and final focus; electronic, electro- and fiber-optics to be utilized in detector readout, accelerator controls and the CCDs required for the vertex detector; as well as high and low temperature superconducting materials (LTSMs) for cavities and some magnets. Our first measurements of fast neutron, stepped doses at the UC Davis McClellan Nuclear Reactor Center (UCD MNRC) were for NdFeB materials at EPAC04[1]. We have extended the doses, included more manufacturer's samples and measured radioactivities. We also added L and HTSMs and various semiconductor and electro-optic materials such as photonic band-gap (PBG) fiber that we studied previously with gamma rays.

INTRODUCTION

This work continues work last reported at PAC05[2] whose goal is to improve systems such as LCs over their lifetimes by providing predictive understanding of radiation damage mechanisms based on more controlled, systematic experiments with neutrons – both fast and slow[3]. Fast neutrons come from bremsstrahlung induced photonuclear reactions and slow neutrons from their moderation.

The UCD MNRC has a number of areas for irradiating samples with neutron fluxes $\leq 4.1{\cdot}10^{10}$ n/cm^{2}s. We used a specialized area (NIF) that allows fast neutron irradiations with 1 MeV equivalent neutron fluxes $\leq 1.5{\cdot}10^{10}$ n/cm^{2}s while greatly suppressing thermal neutrons and γs. We have irradiated individual PM blocks, magnets and a growing array of other materials there as described in [2]. Below, we describe our specific use of the reactor, the measurements and our latest results where we have more than doubled the integrated dose.

LOGISTICAL AND OTHER PROBLEMS

In reviewing experiments in this area, common characteristics emerge that explain both the difficulty and scarceness of systematic, controlled experiments [3]. In fairness, even a brief consideration of such a program shows many questionable and hard to control circumstances such as the difficulty of handling and measuring PM materials even when they are not radioactive. Altogether, this implies that a considerable number of people and jurisdictions become involved. Thus, there is ample opportunity to damage the blocks or change their magnetic properties in ways totally unrelated to radiation damage e.g. most frisking detectors or monitors have steel components that can easily lead to chipped or broken blocks.

While there are many problems, there are many uses for these results. Their importance for accelerators is obvious but there are also opportunities in space applications and materials research such as defect and domain manipulation e.g. whether some forms of damage may be used to improve materials following remagnetization. However, because most new materials tend to be more complex, there are greater possibilities for radiation effects as we discuss for HTSMs such as the BiSCCO compounds.

MAGNETIC MATERIALS

Choice of PM Blocks & Magnets

Most experiments use unloaded, single blocks whose results are difficult to interpret[3]. In magnets, PM dipoles should be less susceptible to damage followed by undulators, wigglers, and magnetic multipoles due to variations in $\vec{M}$ over different block types *and* especially variations in $\vec{H}_{ext}$[4]. This is clear from Fig. 2 of [3] but especially Fig. 4 of [4] and explains our design of an asymmetric quadrupole with simple dipole geometry – shown in [3] for a large gap $G \geq l_x, l_y, l_z$ where it was discussed in detail, together with measurements. All of our blocks are nickel plated but were undoped $Nd_2Fe_{14}B$ there as opposed to those in Table 1 and discussed here.

Table 1: Initial characteristics of open-circuit blocks.

Block	$\mathbf{B}_r$[kG]	$\mathbf{H}_{ic}$[kOe]	$\mathbf{H}_{bc}$[kOe]	$\mathbf{BH}_{max}$
N34Z	11.10	30	10.8	30.8
N50M	13.71	14.9	13.1	47.1
HS36EH	11.60	24.6	11.1	32.8
HS46AH	13.34	13.6	12.5	43.0

Table 1 lists the blocks that were open-circuit irradiated. Figure 1 of Ref. [1] gave typical demagnetization curves from Shin-Etsu for block types N50M and N34Z. All blocks had l_z=6 mm, l_x=9 and l_y=25.4 mm with weights of 10.3 g. These dimensions allowed good uniformity of dose throughout the volume by passing the flux perpendicular to the long dimension[2, 1].

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Radiation Monitoring

Several methods were used to control and monitor the radiation dose and temperatures of the PM materials. First, we sequentially increased the power level from 350 kW up to 1 MW at the reactor and monitored the heating and dose rate over runs of different lengths to reach our current rate of 1 MW for 67 min that keeps the radioactivity at acceptable levels. The containment vessel was rotated with a six-sided holder to isolate the magnets from one another and provide dose uniformity. Initially, we used neutron/photon dosimeter pairs consisting of a PIN diode for the neutron dosimetry that is orders of magnitude more sensitive to neutrons than gammas and MOSFET photon dosimeters where the reverse situation obtains[5]. Also, two sulphur tablets were included whose radioactivity was measured to determine average fluence for every run.

Table 2: Demagnetization in G/Gy for the Table 1 blocks.

Block #	$<\mathbf{M}_y>$[G]	$\mathbf{M}_x^i$[T]±[G]	$-\delta\mathbf{M}_x/\delta\mathbf{D}\|_{\mathrm{MaxD}}$
HS36EH	-35±36	1.1600±1.0	0.00±0.05
HS46AH	-128±20	1.3340±2.0	4.65±0.08
N34Z1	-266±120	1.1091±1.8	0.11±0.02
N50M1	-17±39	1.3707±1.1	2.28±0.05

Magnetic Measurements

For the individual blocks and magnets, a Hall probe fixture was made for field scans in combination with Helmholtz magnetization measurements. Examples were given in Fig. 3 and Tables 1–2 of [3]. Here, Table 2 gives results for the blocks in Table 1 where $<M_y>$ is the average over all runs of a weak component of **M**, x is the easy axis direction and $\mathbf{M}_x^i$ is the measurement before irradiation. The strength errors are small and repeatable. The differential damage $\delta\mathbf{M}_x/\delta\mathbf{D}$ is in G/Gy evaluated at the endpoint of each sequence in Fig. 1. The Hitachi blocks have seen 7 doses totaling 184 Gy(Si) of 1 MeV equivalent neutrons. The Shin-Etsu blocks have seen 11 doses totaling 220 Gy(Si). The damage is roughly linear but shows a progressive (nonlinear) roll-off with dose. Stepped doses are continuing with a goal of 10^{15}n/cm^2.

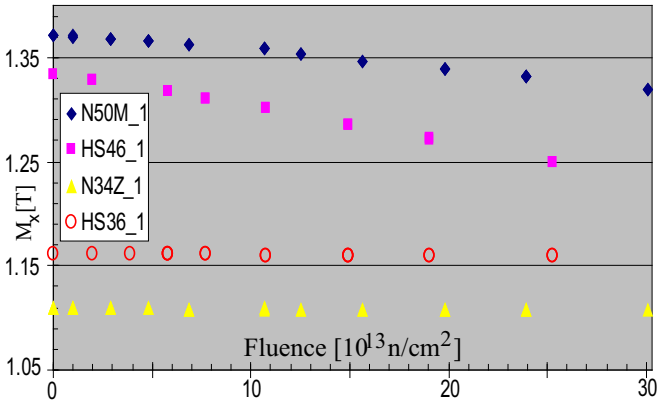

Figure 1: Comparison of nominal 32 and 45 MGOe blocks from Shin-Etsu and Hitachi Magnetics in Tables 1 & 2.

RADIOACTIVITY STUDIES

In this ES&H era, it is surprising that radiation damage studies seldom survey the induced radioactivity but focus on the operational losses even when this seriously affects their logistics and efficiency. Radioactivity has serious short and long term safety implications, is extremely important for the device or system design, development and handling processes and also involves some interesting materials science. While we expect more complex compounds to be less resistant to radiation for several reasons there is one important caveat. Substitutions of like-kind elements such as among transition metals or rare earths in magnetic materials play many roles and are often exceptions.

Magnetic Materials

We know that binary Sm_xCo_y compounds are more radiation resistant and have better thermal stability than those of NdFeB but they also tend to be weaker, more expensive and can become quite radioactive through the large neutron capture cross sections going to the long-lived isotopes Co^{60} [6] and Sm^{151} with 5.3 and 93 y lifetimes.

In contrast, NdFeB is generally cheaper and stronger but less radiation resistant with characteristics that are not as well understood especially when doped with other rare earth substitutions to give $Nd_{2-x}Fe_{14}B$ where x represents other rare-earths such as Dy, Gd, Pr or Tb. Such candidates are seldom considered from this viewpoint but one needs to consider this effect because certain stable isotopes of these have large capture cross sections. Such exotic compounds tend to be proprietary and changing e.g. Shin-Etsu considers N34Z to be a 5$^{\mathrm{th}}$ generation material – still under development. N50M is described similarly as 6$^{\mathrm{th}}$ generation. Both compositions are unpublished but can be inferred from radioactivity measurements. Previous studies[7] have shown such substitutions may improve H_{ic} with a high linear correlation of 0.96 as well as radiation resistance (RR) with a correlation of 0.87 and thus indicate a good correlation of 0.78 between RR and H_{ic}.

$Nd_2Fe_{14}B$ is 26.7% Nd by atomic weight, Fe 72.3% and Boron 1%. While Boron is only 1 %, different models[7, 8] suggest it is the major factor in demagnetization. It has two stable isotopes (A=10 & 11) with $_5B^{10}$ the worst because of its lighter mass and large capture cross section (3.8 kb compared to Co^{59} with 36.6 b) giving it *two* mechanisms for demagnetization. $_{64}Gd^{157,155}$ has 242,61 kb and $_{66}Dy^{164}$ has 3 kb cross sections. A major difference is that B^{10} neutron induced fission can lead to permanent demagnetization while the others lead to similar stable elements.

Radioactivity Measurements Table 2 of Ref. [1] gave trace elements, sources and levels of radioactivity for three different PMs. One of these, "Ref" referred to a 3-block magnet with a thin iron return yoke. Because the overall volumes of material and their geometries were similar, the results can be compared directly. The lower radioactivity in "Ref" was due to use of *undoped* NdFeB in an Fe yoke.

Table 3 gives our results for the samples in Tables 1&2 from Shin-Etsu and Hitachi. The Hitachi blocks are doped differently using Pr^{141} and Dy with 7 stable isotopes versus Tb^{159} for Shin-Etsu. The differences are compounded by Shin-Etsu's partial use of Co in place of Fe. This Table comes from Ref. [2] and includes only the strongest line from each of the strongest isotopes that were observed.

Superconducting Materials

Current leads, conductors and cavities with minimal Joule losses are highly desirable – especially if options exist for low thermal conductivity. HTSMs such as $Bi_2Sr_2Ca_2Cu_3O_5$ are also interesting because they are quite complicated and share several characteristics with NdFeB being crystalline, complicated and accommodative to substitutions. Since their primary functional characteristics are not understood, perturbative damage mechanisms can provide useful insights. Because LHC neutron fluences of order 10^{15}/cm^2 are comparable to our goal for other samples, we decided to look at these materials to see whether a useful program was possible[9].

BSCCO samples were from American Superconductor and Sumitomo with comparable amounts of Ag constituting about 60 % of the 4 mm by 0.3 mm tapes. Typical LTSMs were Formvar coated, $Nb_{53}Ti_{47}$ with 0.5 mm by 0.7 mm. All samples were capable of order 80-90 A at 1 μV/cm. A test setup was made for this current range while irradiating small test samples. A 5 cm length of BSCCO (< 0.5% of the overall mass) totally dominated the radiation level of the combined package. A day after irradiation with $4{\cdot}10^{13}$n/cm^2, it had 0.93 R/hr β and 0.13 R/hr γ on contact. With Ag being 48% Ag^{109}, it is not surprising to see Ag^{110m} excited strongly with many high energy γs up to 1.5 MeV and activities of a μCi/10^{13}n/cm^2. Many other elements were observed with varying degrees of confidence including Co, Cr, Fe, Mn, Nb, Ru, Sr, and Zn. Many came from the stainless steel cladding. Several other, comparatively strong lines have yet to be identified.

INTERPRETATION AND DISCUSSION

All PM blocks were doped differently. Shin Etsu clearly made significant substitutions of Co for Fe by the relative intensities of Co and Mn in the different blocks. Also, there were many strong Tb^{160} levels from n-capture on Tb^{159}. Based on known capture cross sections for Fe^{58} and Tb^{159} and their relative abundances there is a large substitution in N34Z that improves RR although HS36 is better while HS46 is most susceptible. Curiously, the Co substitutions appeared to help neither strength nor radiation resistance.

Considering the amount of iron, it is interesting that so few lines appear e.g. Mn^{54} from Fe^{54}(n,p) charge-exchange having a rather large cross section but a threshold of a few MeV. Mn^{56} appears in a similar way from Fe^{56} but with an even lower intensity and 2 h lifetime. From all of this, NdFeB has advantages over SmCo from both the lifetimes, γ energies and relative intensities.

Table 3: Radioactivities and half-lives of species in Table 2.

Element $_Z X^A$	Half Lives	Energy [keV]	Radioactivity[μCi]			
			N34Z	N50M	HS36	HS46
$_{65}Tb^{160}$	72.3d	298.6	0.83	0.44	-	-
$_{27}Co^{60}$	5.27y	1332.4	0.08	0.06	-	-
$_{61}Pm^{151}$	28.4h	340.1	0.40	0.77	0.53	0.78
$_{25}Mn^{54}$	313d	834.8	0.01	0.01	0.09	0.18
$_{59}Pr^{142}$	19.2h	1575.6	0.01	0.01	0.20	0.29
$_{61}Pm^{149}$	53.1h	285.9	0.32	0.50	0.36	0.50
$_{66}Dy^{165}$	2.33h	94.8	-	-	0.19	0.03
$_{60}Nd^{147}$	11.0d	91.2	0.02	0.02	0.02	0.02

We hope to see whether Oxygen plays a similar role in HTSMs to Boron in PMs by including $Bi_2Sr_2Ca_1Cu_2O_y$ because little has been done with light element substitutions but have not yet figured out how to deal with the high radioactivities in an efficient way.

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[9] Sample materials were kindly supplied to us by Ramesh Gupta and Bill Sampson at BNL and Steve St Lorant at SLAC.